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The research objectives of this program have focused on the development of blue/green light emitters. The effort was initially based on the II-VI compound semiconductors, but was later directed at the group III nitrides. As a result of the changes in objective, the summary of research activity addressed by this report involved the growth of both widegap II-VI and group III nitride materials as well as device studies in both.

In the second part of the program we studied the growth and the optical evaluation of wise bandgap nitride heterostructures, an effort which included the first reporting of a GaN-based laser to be fabricated (in collaboration between Brown and the Cree Corporation) in the US, and the first to be grown on SiC substrates.

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II-VI/III-V Heterojunction Lasers
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Objectives

The research objectives of this program have focused on the development of blue/green light emitters. The effort was initially based on the II-VI compound semiconductors, but was later directed at the group III nitrides. As a result of the changes in objective, the summary of research activity addressed by this report involved the growth of both widegap II-VI and group III nitride materials as well as device studies in both.

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Wide Bandgap II-VI Light Emitters - II-VI semiconductors

For several years a key aspect of our AFOSR research program had been the study of the heterovalent interface, specifically the ZnSe/GaAs interface. This interface became crucial with the need to increase the operating lifetime of the blue/green laser diodes. It appeared that the laser device failure was primarily the result of extended defects, such as stacking faults, originating at, or near the ZnSe GaAs interface, and producing threading dislocations which pass through the quantum well gain region of the device. TEM studies have shown that the point where the threading dislocation passes through the quantum well acts as a nucleation point for a network of dislocation loops forming dark patches where the gain is greatly reduced, ultimately ceasing the lasing action. At the beginning of the study, it was not at all clear to what extent the generation of extended defects at the II-VI/III-V interface was an intrinsic property of the interface, in which case the density could not be significantly reduced. The density of extended defects was initially often in the $10^7 cm^{-2}$ range. As efforts to reduce the density progressed, densities in the 105cm-2 were often observed, but inconsistently. It gradually became clear the high densities were not intrinsic to the nucleation process. The question then was how low a density could one achieve. We were finally able to reduce the density of extended defects into the $10^3 cm^{-2}$ range, and most importantly, the results were quite consistent from run to run. At this point it actually appeared that the density of extended defects was a function of the quality of our GaAs epilayer, and not related to the heterovalent nucleation process. The impact on device performance was significant. Although the complications of laser fabrication tended to obscure the improvement, the operation of LED devices provided clear evidence that the situation was quite different in device structures having the low density of extended defects. In a lifetime study, LED devices were operated at $100 A/cm^{-2}$, a level compatible with laser threshold values. The density of extended defects was sufficiently low that, for the first time, it was possible for many devices to be free of the extended defects which had heretofore resulted in the growth of dark patches in the active region. The LED devices in this study exhibited lifetimes of 500 hours before failure.

Group III-nitrides for Wide Bandgap Light Emitters

The Brown/Purdue effort in the nitrides began in February 1996 with the installation of the appropriate source cells in the III-V growth chamber of the modular MBE. Nitrogen was supplied either in the form of uncracked ammonia, from an electron cyclotron source, or from a liquid nitrogen cooled high power rf plasma source. Most of the experiments to date have been conducted with the rf plasma source. The effort began in collaboration with Hewlett-Packard who supplied us with MOCVD-grown (on sapphire) GaN epilayers employed in lieu of direct substrates for subsequent MBE growth. Later on, we were supplied by GaN epilayers from Sandia National Laboratories. An objective was the exploration of means for improving the quality of quantum well structures in the nitride system.

An additional objective is the exploration of means for improving the quality and the growth of InN by MBE. Both the GaN and InN films were grown and evaluated by PL, XRD rocking curves, crossectional TEM, and RHEED. One overall issue we have addressed is the lack of hard temperature data in the literature. The mounting of samples with In does not work due to the loss of the In at the temperatures associated with GaN growth. We have found some success with a "glue" composed of equal parts of In and Sn except for the very highest temperatures of growth. It is important to correlate flux ratios with actual substrate temperatures as the growth conditions are very much a function of temperature.

(1) Growth of InN

The interest in the growth of InN partially stems from the potential for InN, or high concentration alloys of (GaIn)N, to be incorporated in heterostructures designed to form low resistance contacts to p-GaN. In addition, although a potentially important member of the group III-nitride family, in fact InN has received relatively little attention. As mentioned above, one reason for the lack of reports in the literature is the growth difficulty arising from the low dissociation temperature of InN, a condition which essentially precludes the use of the temperatures associated with MOCVD. Even for MBE there is a growth challenge as InN exhibits significant dissociation at temperatures even below 400°C.

During the course of research we have employed a quadrapole mass analyzer placed in a source position on the MBE source flange in order to obtain additional insights into the kinetic processes occurring during the growth of InN. An important point is the manner in which the dissociation of InN leaves behind In, which in turn has a relatively low vacuum evaporation rate such that there tends to be an accumulation of In. Further, while InAs and InSb do not present serious problems for MBE growth, InN differs from these compounds in having a relatively low heat of formation. As a result of this, as well as the possibility of there being kinetic barriers present, In is not as reactive with N as it is with As or Sb. Herein lies a problem in the sense that it appears that In is "as likely" to bond to another indium atom (forming In clusters) as it is to form InN.

(2) Studies of In incorporation into GaN and AlN.

In the growth of GaAs and alloys it is traditional to assume that all the metal species exhibit unity sticking coefficients, a feature which greatly simplifies the tuning of flux to achieve a particular alloy composition. One issue which has received little attention for the group III nitrides concerns the actual sticking coefficients of the alloy constituents as functions of substrate temperature, flux ratio and atomic nitrogen flux; the metal sticking coefficients are not unity in general. We have explored this parameter space using real-time acquisition of RHEED data to observe the lattice parameter relaxation, and hence changes in lattice spacing as a measure of alloy fraction. Such experiments have been performed with both (Al,In)N and (GaIn)N. Interest in the AlInN alloy stems from our interaction with Sandia where there is interest in exploring the UV transitions in GaN quantum wells bounded by lattice-matched (Al,In)N barrier layers. The (Ga,In)N alloy is of interest for both heterostructure contact applications as well as the importance of the alloy in the active region of laser diodes.

(3) MBE growth of GaN

We have been employing real time RHEED studies to explore the growth space for GaN. Despite the considerable effort already expended in various laboratories for the growth of GaN, the interplay of the various parameters on the growth are not well understood. The growth is complicated by the twin processes of Ga desorption and compound dissociation at the higher growth temperatures, and the tendency for Ga saturation and accumulation at lower temperatures. Of interest is the relationship between the various parameters having impact on the "quality" of the resultant film. A computerized RHEED set-up with CCD camera and computer processing of the real time data was used for these experiments. In principal growth rates are readily obtained in situ by RHEED oscillations. However we have found that there is not necessarily a one-to-one correspondence between an oscillation period and the growth of a single monolayer.

(4) Brown contribution to nitride work

In the initial phases of the nitride research, the group at Brown provided a comprehensive battery of optical techniques to evaluate the materials grown at Purdue. Additionally, initial experiments have been conducted to study the optical properties of GaN-based heterostructures, especially under high elecron-hole pair injection. This subject is still in its infancy but is crucial to the understanding of the formation of optical gain e.g. in InGaN quantum wells. The present state of the art shows very broad optical linewidths associated with the luminescence, reflectance, of absorption spectra in InGaN, even at relatively modest In-concentration (10%). Our studies, which include materials grown at Purdue, Hewlett-Packard, as well as commercial Nichias LEDs, strongly suggest that the principal QW optical resonance is strongly influenced by crystal disorder, including large potential energy fluctuations on a microscopic scale. For example, we have applied time-resolved spectroscopy to measure surprisingly long e-h pair radiative lifetimes which, furthermore, show a distinct dependence on the photon energy of emission [1]. Such behavior

indicates a highly inhomogeneously broadened resonance which may correspond in the real space to nanoscale clusters of InGaN within the 'quantum well'.

In collaboration with Sandia National Laboratories, the group at Brown has focused on the design and fabrication of ultraviolet GaN quantum well light emitting diodes [2]. First devices have been demonstrated at the shortest reported wavelength in the nitride emitters to date. The room-temperature electroluminescence emission was peaked at 353.6 nm with a narrow linewidth of 5.8 nm. In the simple planar devices, without any efforts to improve light extraction efficiency, an output power of 13 μW at 20 mA was measured, limited in the present design by absorption in the GaN cap layer and buffer layer. Pulsed electroluminescence data demonstrate that the output power does not saturate up to current densities approaching 9 kA/cm2.

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